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NEAR-OPTIMUM PROCEDURE FOR HALF-LIFE MEASUREMENT BY HIGH-RESOLUTION GAMMA-RAY SPECTROMETRY

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A near-optimum procedure for using high-resolution γ-ray spectrometry to measure the half-lives of appropriate γ-ray-emitting-nuclides is presented. Among the important points of the procedure are the employment of the reference source method for implicit correction of pileup and deadtime losses; the use of full-energy peak-area ratios as the fundamental measured quantities; and continuous, high-rate data acquisition to obtain good results in a fraction of a half-life if desired. Equations are given for estimating the precision of the computed half-lives in terms of total measurement time, number of spectral acquisitions, and the precision of peak-area ratios. Results of ^{169}Yb half-life measurements are given as an example of the procedure's application.

1. Introduction

High-resolution γ-ray spectrometry with germanium detectors is now frequently used to refine previously measured half-life values for appropriate γ-ray-emitting nuclides. It appears from recently published work that some of the power and advantages of automated high-resolution systems are often neglected in half-life measurements. Therefore, it seems useful to offer a few suggestions to efficiently use high-resolution γ-ray spectrometry for such measurements. The suggestions, taken together, constitute what is thought to be a near optimum procedure; most of them have been used before in some form but seem not to have been used together as an integrated procedure. They are particularly useful for obtaining reasonably precise and unbiased half-life values in a fraction of a half life but should also be helpful in improving both the accuracy and precision of measured half-life values in general.

2. Advantages and Use of the Reference Source Method

Data acquisition with high-resolution germanium detectors suffers rather large losses from deadtime and pileup. The accurate correction of those losses has long been a vexing problem in the application of high-resolution γ-ray spectrometry to quantitative problems, especially at high input rates. However, the reference source method seems to offer advantages over any currently used electronic or computational method.

By reference source we mean a γ ray source geometrically fixed with respect to the detector, to the source being measured, or both, and used to make corrections either implicit or

explicit–for deadtime and pileup. The reference source method (as well as most, if not all, electronic methods) is based upon the assumption that all the full-energy peaks in a high-resolution γ-ray spectrum suffer the same fraction of loss because of the combined electronic effects of pileup and deadtime. By observing appropriate precautions, the loss fraction can indeed be made very nearly equal for all peaks, especially over narrow energy ranges. Thus, the ratios of net full-energy peak areas (assuming fractional losses constant throughout the counting interval) are independent of deadtime and pileup losses, as well as of count times. Ratios of peak areas from the measured nuclide and the reference source become the measured quantities, rather than absolute counting rates. If a system is properly set up, one does not have to calculate explicit corrections for deadtime and pileup.

Of the precautions observed to equalize the loss fractions to the two spectral peaks of the measured ratios, the most obvious is to use γ-ray pairs with small energy differences, thereby minimizing any small problems relating to the variation of peak width with energy or to the "magnitude" of pileup as a function of energy. Additionally, the electronics can usually be adjusted to minimize the variation of peak width with both energy and count rate. Shorter time constants–which reduce the fraction of loss that must be corrected–and high base-line restorer settings often help to minimize those variations. Pileup rejection (PUR) also helps preserve peak width and shape, especially when high rates are employed, and need not, as is sometimes feared, actually cause variations in the fractions of loss.

Other precautions involve the accumulated pulse-height spectrum. Peaks that have adequate ratios of net area to the Compton continuum area beneath the peak (≥10:1 if possible) should be employed for the measured ratios to minimize effects of any curvature of the Compton continuum in the peaks' regions. Finally, and importantly, peaks should be avoided in regions where the Compton continuum has significant curvature, Compton edges, or random sum peaks.

For half-life measurements, the reference source will usually be spatially fixed relative to the source of the measured nuclide, either by actually mixing the two source materials or by firmly fastening two small, discrete sources as close together as possible. The position of the combined source will ordinarily be fixed relative to the detector as well, however, if there are small variations in the source-detector distance, $1/r^2$ effects on the relative counting rates will be essentially eliminated by the use of the combined source, and the magnitude of the measured ratios will be unchanged. Additionally, the combined source should preferably have sufficient intensity to get adequate counting rates when placed ≥10 times the detector diameter from the detector. Any residual $1/r^2$ effects, either from changing count rates or from changing probabilities of cascade summing, are thereby minimized.

If the logarithms of the measured ratios are plotted against the measurement times and are fitted by standard methods to a straight line, the slope of the line is and the decay constants of the nuclide of uncertain half-life and the reference source, λ_U and λ_R, are related by

$$\lambda_u = -m + \lambda_r \quad (1)$$

The half-life T_u is, of course, obtained from $T_u = \ln 2/\lambda_u$.

The error caused in λ_u by the error in λ_r is given by

$$(d\,\lambda_u/\lambda_u) = (\lambda_r/\lambda_u)(d\,\lambda_r/\lambda_r) \quad (2)$$

Obviously, it is desirable for the half life of the reference source to be greater than that of the measured nuclide so that the factor (λ_r/λ_u) will be less than one. Usually reference sources can be found for which the estimated error in λ_r is so small that, when multiplied by this factor, the influence on λ_u is negligible.

3. Half-Life Precision and Total Measurement Time

Automated multichannel-analyzer systems make possible almost continuous data acquisition, and if it is done at high throughputs (implying high input rates), it is possible to obtain quite precise values for half-life measurements in only a fraction of a half-life. There are even some advantages in shorter measurements: the probability of equipment failure during measurement is minimized, and equipment utilization is maximized. Problems caused by changing count rates, equipment instabilities, or drifting detector efficiencies are minimized.

We will consider the precision of a half-life value based on only two measured ratios and then based on many measured ratios. But first, a remark on precision and its notation is required. We shall refer to the relative standard deviation (RSD) of the measured ratios as predicted from the information extracted from a single high-resolution spectrum and also to the predicted RSD of quantities calculated from those measured ratios and predicted RSDs thereof. We adopt the Roman letter u to represent the predicted RSD; this is in contrast to the sample RSD, generally called the coefficient of variance, usually denoted by the Roman letter v. We emphasize that when properly formulated, the predicted RSDs are generally more accurate as estimates of population parameters than sample RSDs computed from relatively small samples. We note that the predicted RSDs are based upon the assumption that the sums of total counts in spectral regions of interest (ROIs) obey Poisson statistics. This assumption is not quite true for systems with significant counting losses, but because any individual ROI usually contains only a small fraction of the counts in the total spectrum, it is almost always adequate.

With this notation, the predicted RSD of the half life, $u(T)$, based upon two measured ratios R_1 and R_2 and their predicted RSDs $u(R_1)$ and $u(R_2)$ is

$$u(T) = (1/\ln 2)(T/\Delta t)\sqrt{u^2(R_1) + u^2(R_2)} \quad , \tag{3}$$

where Δt is the interval between the starts of the two ratio measurements. If $u(R_1) \sim u(R_2) = u(R)$, which is usually possible, the expression becomes

$$u(T) = (\sqrt{2}/\ln 2)(T/\Delta t)\, u(R) \quad , \tag{4}$$

where $\sqrt{2}/\ln 2 = 2.04$.

We next consider the estimated precision of half-life values computed from many ratio measurements. Assume n ratio measurements, let the n count times be equal, let them begin at equal intervals, and let the total interval between the beginning of the first and the beginning of the last counts be Δt. Assume also that the predicted RSDs of all the ratios are approximately equal and indicated by u(R). It can then be shown that

$$u(T) = (2\sqrt{3}/\ln 2)(T/\Delta t)\{[(n-1)/n]\sqrt{[n/(n^2-1)]}\}\, u(R) \tag{5}$$

For n = 2, Eq. (5) reduces to Eq. (4) as required, and we note that $[2\sqrt{3}/\ln 2] = 4.998$. As before, u(T) is proportional to $T/\Delta t$. Now define $F(n) = \{[(n-1)/n]\sqrt{[n/(n^2-1)]}\}$, the function of n that shows the improvement in estimated precision as the number of ratio measurements is increased. Table 1 gives values of F(n) and F(n)/F(2) for a selection of values of n. The values of F(n)/F(2) indicate the improvement of the precision of the n-point measurements over the 2 point measurements. Note that the function F(n) has the same value for n = 2 and for n = 3, showing that three equally spaced ratio measurements are no better than two for computing a half life value, and note also that for $n >> 1$, $F(n) \sim 1/\sqrt{n}$.

If the counting is essentially continuous then the length t of the n counting intervals is $t = \Delta t(n-1)/n$. The estimated RSDs of the ratios u(R) are, if properly calculated, proportional to $1/\sqrt{t}$. For $n >> 1$, we then find that

$$u(T) \sim 1/(n\,t)^{1/2} \tag{6}$$

This result indicates that for a fixed total time, u(T) does not depend upon the number of intervals into which that time is divided, and that for a nonfixed total time, u(T) is proportional to both $1/n^{1/2}$ and $1/t^{1/2}$.

Consider the ratio measurement time t. It has been shown that if fractional counting losses are constant during the interval t, it does not matter what fraction t is of T. It is obvious that if the interval t is any significant fraction of T, the counting losses will certainly not be strictly

Table 1
Values of the Precision Function F(n) and Ratios F(n)/F(2) for Different Values of n

n	F(n)	F(n)/F(2)
2	0.408300	1.00000
3	0.408300	1.00000
4	0.387300	0.94900
5	0.365200	0.89400
10	0.286000	0.70100
20	0.212700	0.52100
22	0.203700	0.49900
100	0.099000	0.24300
200	0.070360	0.17200
500	0.044630	0.10900
598	0.040820	0.09999
1 000	0.031590	0.07740
10 000	0.009999	0.02456

constant over the interval. Computational study shows that the reference source method produces half-life results with small or negligible error for reasonable values of t. For $t \leq 0.01$ T the error is less than 1 part in 10^6 for all ordinary conditions. Even for $t = 0.1$ T, the error is rarely as great as 1 part in 10^3.

If the best possible precision of the measured half life is to be obtained in a fixed time, the initial total input rate should give near maximum output into the stored spectra. The throughput curve can usually be experimentally determined in a short time for any data acquisition system or, in some cases, can be estimated with sufficient accuracy. In many modern γ-ray spectrometry systems, the analog-to-digital converters (ADC) are so fast that for some amplifier time constant settings, the conversion and storage are complete before the amplifier output has returned to a level low enough for the ADC to accept another event. In such circumstances, the ADC's do not contribute to the counting losses at all--pileup is the only loss mechanism--and the throughput of the system is well approximated as an ideal paralyzable system with a fixed deadtime dt. For such a system, the stored rate, or throughput, sr is given in terms of the total rate r and dt by the well known expression

$$sr = r e^{-r\,dt}, \qquad (2)$$

where r is the time dependent gross count rate from the detector. This curve peaks at $r = 1/dt$, and has a broad maximum such that about 90% of maximum throughput is achieved for $r = 0.6/dt$. Ordinarily, the initial input rate might be adjusted to give about 90% of maximum throughput, with any additional throughput obtained at the price of considerably higher input rates with accompanying degradation of both peak width and shape. Exact dt values will vary with the details of the electronic acquisition system being used, but for systems

employing ordinary spectroscopy amplifiers with semi-Gaussian shaping and ADCs requiring the amplifier output to drop below the input discriminator level to accept an event, it is about nine times the shaping-time constant selected.

4. Full-Energy Peak Areas, PUR, and Peak Stabilization

It is important to obtain correct estimates of the RSDs of the full-energy peak areas to properly weight the area ratios in the least-squares fitting procedures and to accurately predict the RSD of the final half-life. If one must use γ-ray peaks, which are part of overlapping spectral multiplets, then using spectral unfolding codes is mandatory. Unfortunately, the precision estimates given by many such codes are not always very accurate, especially when the number of counts in the peaks is large ($\geq 10^5$), but will usually suffice. When it is possible to use well-resolved spectral peaks, ROI methods give peak-area values as accurate as any method, and the precision estimates are correct for any number of peak counts. Expressions for both the net peak areas and their predicted RSDs are given elsewhere [1] for one successful implementation of the ROI scheme.

Using PUR to acquire spectral data is desirable because it improves both peak width and shape, thus making the extraction of full-energy peak areas easier and more accurate. Occasionally concern is expressed about employing PUR while acquiring half-life measurement data because it is thought the PUR might change the full-energy storage fractions as a function of energy. There are two types of PUR in common use, only one of which should cause concern. In the safer method, an event is denied storage only when the electronic criteria for piled up events are met. In the less-safe method, any stored event must meet the prescribed electronic criteria for <u>not</u> being piled up. In PUR systems of the latter type, all accepted events must trigger a low-threshold fast discriminator. In some such systems, it has been observed that the discriminator thresholds "roll off" at lower energies in such a way that the fraction of events triggering the discriminator is a function of energy and, even worse, of time (probably because of temperature or aging effects). PUR schemes using that kind of logic should be carefully tested before use in applications for which long term stability and high precision are required.

Digital stabilizers are now available that can constrain the centroids of two selected spectral peaks to specified channels. With the positions of two peaks fixed, the centroids of none of the spectral peaks should ever drift by more than a tenth of a channel. Such stabilization is advantageous for long counting periods, especially when using fixed ROIs to obtain the peak areas because the positions of the full energy peaks must be fixed within the ROIs selected.

5. Example: The Half-Life of ^{169}Yb

The remeasurement of the ^{169}Yb half-life illustrates the use of the above suggestions, most of which were formulated while nondestructive assay (NDA) systems were developed to measure uranium and plutonium nuclides. Ytterbium-169 is often used in the assay of ^{235}U, and, in 1983, there seemed to be enough uncertainty in its half life (which is required in the long-term operation of the NDA systems) to justify a remeasurement. Fortuitously, ^{235}U was an ideal reference source for measuring the half-life of ^{169}Yb.

The two most intense γ-rays of ^{169}Yb are 177.2 keV and 198.0 keV. The 198.0-keV γ-ray suffers slight interference from some of the weaker γ-rays of ^{235}U, so the 177.2 keV γ-ray was used for the half-life measurement and the principle γ-ray of ^{235}U (185.7 keV) was used as the reference. Both are clean, well-resolved peaks with an energy difference of only 8.5 keV. A combined source was made by fastening a ^{169}Yb source directly to the surface of a nickel-plated, highly enriched uranium disk. At a distance of about 15 cm from the end cap of a 10% efficient Ge(Li) γ-ray detector, the combination source gave a gross rate of about 33 000 s^{-1}, which gave about 90% of the maximum throughput of the acquisition system. At the beginning of the exercise, the rate of the 177.2-keV γ-rays was about 20% greater than that of the 185.7-keV γ-rays.

A standard data acquisition system was used, which included a 100-MHz Wilkinson-type ADC and two-point digital stabilization. Only 1 024 channel spectra were used in order to minimize deadtime losses, which were still significant with the relatively slow ADC employed. The amplifier was used with 1-μs time constants to minimize pileup losses, and the built-in PUR (of the safer type described above) was used.

ROI methods extracted the full-energy peak areas and computed their predicted RSDs. The midpoint elapsed time, the ratio of the unknown to reference peak areas, and the predicted RSD of the ratio were stored for each count. The half-life values were computed as described above using standard weighted least squares methods [2].

Seven values of the half-life were obtained from seven data sets. Acquisition was on weekends or holidays when equipment was not otherwise in use, thereby reducing data acquisition periods to 3.5 days or less. The details of the acquisitions and resulting values are given in Table 2. As seen, the total acquisition times were all about one tenth of the ^{169}Yb half life, with predicted %RSDs between 0.32% and 0.44%. The weighted and unweighted averages of the seven measured values agree to better than 1 part in 3 000, and the sample and predicted RSDs of the averages agree to two significant figures. The agreement of the values while gratifying, is fortuitous because an RSD value based on a sample of seven values must itself have an estimated RSD of ~30%. The reduced chi square statistic values and examination of data plots indicate that the assumed relationship accurately describes the measured data. In all

Table 2
Measurements of Half-Life of ^{169}Yb

No. of Spectra	True Time/ Spectrum (s)	Meas. Period (HL Frac)*	Measured Half-life (d)	Predicted RSD (%)	Reduced Chi-Square
500	600	0.110	31.75	0.38	0.92
500	600	0.110	31.96	0.35	0.91
500	500	0.093	31.98	0.37	0.99
500	500	0.093	31.77	0.37	1.12
462	500	0.086	31.76	0.42	1.00
500	600	0.110	31.89	0.32	0.99
416	600	0.092	32.04	0.44	1.01
Unweighted Av and Sample RSD(%)			31.88	0.37	
Sample RSD(%) of Av				0.14	
Weighted Av and Predicted RSD(%)			31.88	0.14	

*HL Frac means half-life fraction.

cases very nearly the expected fractions of points are within one, two, and three predicted standard deviations of the fitted line. There is no obvious evidence in Table 2 or the data plots to indicate significant bias in the final value of the ^{169}Yb half-life of 31.88 d with an estimated RSD of 0.14%. However, because this value differs from that recently published in NCRP Report No. 58 by about 0.5% [3], another measurement using somewhat longer acquisition periods is planned in the near future.

In closing, we give two examples (from the sixth data set) of how Eq. (5) can estimate the expected precision from a data set. For that set of data, $(T/\Delta t) \sim 9.1$, and $F(500) = 0.0446$. The values of $u(R)$ ranged from 0.1562% to 0.1601% with an average of 0.158%. Using these values in Eq. (5), we get $u(T) \sim (5)(9.1)(0.045)(0.158\%) \sim 0.32\%$, which agrees with the value generated from the weighted least-squares fitting of the data. Finally, it is interesting to predict the precision of the measured half-life if the measurement had occupied about one-half of the half-life (500 counts of 3 000 s each rather than 500 counts of 600 s). We would then have $u(T) \sim (5)(2)(0.045)(0.16/\sqrt{5}) \sim 0.03\%$, about a factor of 10 better.

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